

Kawasaki, Araki and Tanaka Reply: In the preceding Comment [1] Sausset and Tarjus (ST) proposed an alternative scenario for the slow dynamics in our two-dimensional (2D) polydisperse colloidal liquids [2], based on the frustration-limited domain theory [3] which focuses on self-generated frustration in the order parameter itself as in [4]. ST claimed that sufficiently polydisperse hexatic order is not space-filling, so it is a 2D analog of the icosahedron. We agree with the former, but the latter seems to be subtle due to the lack of the uniformity of frustration. An exact 2D analog may be hexatic ordering on a surface of incommensurate constant curvature in the sense that in both cases frustration is ‘uniform’ [3, 4]. We regard the same phenomenon as random-field effects on (quasi-)long-range crystalline ordering [2, 5, 6]. Since we described our thoughts on the differences between the two approaches in detail in [6], we do not repeat it here.

First we show the analysis proposed by ST in Fig. 1(a). Their function $L^* \sim B[(\phi - \phi_I)/\phi_I]^x + C$ fits reasonably well to our data. Here ϕ is the volume fraction of colloids and ϕ_I is ϕ at the hexatic ordering for polydispersity $\Delta = 0\%$ ($\phi_I \sim 0.57$) [Fig. 1(b)]. The fitting yields $x \sim 3$, consistent with the suggestion of ST [1]. x is suggested to be related to the correlation length exponent of the unfrustrated system [3]: For the present case (2D hexatic ordering), $\xi_6 \sim e^{h[(\phi_I - \phi)/\phi]^{-1/2}}$ [4]. The physical meaning of $x \sim 3$ needs to be clarified along this line. Since our model predicts the divergence of ξ toward ϕ_0 whereas their model predicts the absence of any such singularity, the difference between the two predictions should more evidently appear near ϕ_0 . So we made simulations at $\phi = 0.64$ for a system of 16384 particles [see the points in the yellow (shaded) circle in Fig. 1(a)]. Unfortunately, the difference is too small to draw any conclusions.

Here we mention the work of Santen and Krauth [8], which demonstrate that there is no ideal glass transition point ρ_G for $\Delta \sim 50\%$ (in our definition of Δ [2]). They determined ρ_G by fitting the diffusivity D by $(\rho_G - \rho)^\alpha$ as $\rho_G = 0.805$. This ρ_G corresponds to $\phi_G = \rho_G/\sqrt[3]{2} = 0.64$

in our notation. This fitting function is not the Vogel-Fulcher type, but that for mode-coupling theory. Thus, ϕ_G is the mode-coupling ϕ_C and we expect that $\phi_0 > \phi_G = \phi_C$. In our opinion, thus, what they demonstrated is that there is no thermodynamic phase transition at the mode coupling ϕ_C . According to our state diagram [Fig. 1(b)], $\phi_G = 0.64$ for $\Delta \sim 50\%$ may be located far below ϕ_0 . Our experiments on 2D driven granular systems [9] also demonstrated that for $\Delta = 10.7\%$ $\rho_0 = 0.838$, which is higher than ρ_G for $\Delta \sim 50\%$ [8].

Next we mention our previous simulation study of a system with competing orderings [7]. In this case, the underlying crystalline order is anti-ferromagnetic and the crystallization is of first order. Nevertheless, we observe behavior very similar to the present case. The basic features of the phase diagram are also very similar between the two [compare Fig. 1(b) with Fig. 2 of [7]]. These facts seem to support our scenario.

Finally, we note that our preliminary study on 3D polydisperse colloidal liquids indicate that there exists medium-range crystalline ordering (fcc or hcp), which is not icosahedral, and $\xi \propto (\phi_0/\phi - 1)^{-2/3}$, consistent with our prediction [5]. This also supports our scenario. At the same time, however, a recent study by Coslovich and Pastore favors the scenario of frustration-limited domain theory [3] rather than ours. Thus, further careful studies are required to settle the issue of the role of frustration in the glass transition. Such efforts will ultimately lead to a clear physical understanding of the glass transition.

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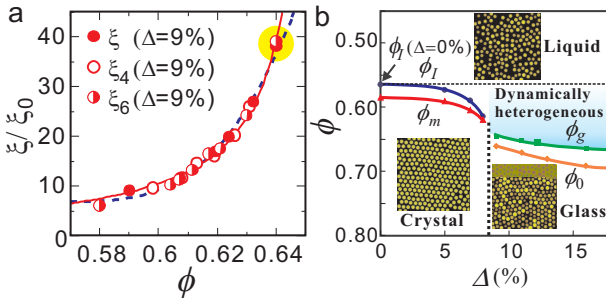


FIG. 1: (color online). (a) ϕ dependence of the scaled correlation length ξ/ξ_0 for $\Delta = 9\%$. Solid and dashed curve are the fittings of $(\phi_0/\phi - 1)^{-1}$ and $(\phi - \phi_I)^3$, respectively. (b) State diagram of polydisperse colloidal liquids in the ϕ - Δ plane.

- [1] F. Sausset and G. Tarjus, Preceding comment.
- [2] T. Kawasaki, T. Araki, and H. Tanaka, Phys. Rev. Lett. **99**, 215701 (2007).
- [3] G. Tarjus, S.A. Kivelson, Z. Nussinov, and P. Viot, J. Phys.: Condens. Matter **17**, R1143 (2005).
- [4] D. R. Nelson, *Defects and Geometry in Condensed Matter Physics* (Cambridge Univ. Press, Cambridge, 2002).
- [5] H. Tanaka, J. Phys.: Condens. Matter **10**, L207 (1998); J. Chem. Phys. **111** 3163, 3175 (1999).
- [6] H. Tanaka, J. Non-Cryst. Solids **351**, 3371 (2005).
- [7] H. Shintani and H. Tanaka, Nature Phys. **2**, 200 (2006).
- [8] L. Santen and W. Krauth, Nature **405**, 550 (2000).
- [9] K. Watanabe and H. Tanaka, submitted.
- [10] D. Coslovich and G. Pastore, J. Chem. Phys. **127**, 124504 (2007).